REVIEW

Nuclear fission: the interplay of science and technology

BY A. M. STONEHAM*

London Centre for Nanotechnology, and Department of Physics and Astronomy, University College London, Gower Street, London WC1E 6BT, UK

When the UK’s Calder Hall nuclear power station was connected to the grid in 1956, the programmes that made this possible involved a powerful combination of basic and applied research. Both the science and the engineering were novel, addressing new and challenging problems. That the last Calder Hall reactor was shut down only in 2003 attests to the success of the work. The strengths of bringing basic science to bear on applications continued to be recognized until the 1980s, when government and management fashions changed. This paper identifies a few of the technology challenges, and shows how novel basic science emerged from them and proved essential in their resolution. Today, as the threat of climate change becomes accepted, it has become clear that there is no credible solution without nuclear energy. The design and construction of new fission reactors will need continuing innovation, with the interplay between the science and technology being a crucial component.

Keywords: materials; fission; nuclear energy; radiation damage; radioactive waste; research organization

1. Introduction

In 1947, the first nuclear reactor in western Europe was switched on, the tiny Graphite Low Energy Experimental Pile (GLEEP) reactor at Harwell, near Oxford, UK. In 1956, the nuclear power station at Calder Hall was connected to the grid and began delivering electricity in commercial quantities, the first such power station in the world. The first reactor was in use for nearly 47 years when the station closed in 2003. These time scales seem incredible today. The technology was new, and much of the science was still being created, yet the building of Calder Hall took only about 3 years. True, there were Cold War pressures that gave priority to the nuclear field. But the quality of the work is evident by the ultimate, demonstrated, reactor lifetime being significantly longer than economists will allow in costing nuclear electricity today.

*a.stoneham@ucl.ac.uk.

One contribution of 13 to a Discussion Meeting Issue ‘Energy materials to combat climate change’.
How was this achieved? This was a case when the best basic and applied science came together with formidable engineering. As Sir Alan Cottrell (Cottrell 1992) wrote, new scientific knowledge was needed for the solution of the practical problems. It was not the mere application of existing knowledge, but research ‘indistinguishable from pure scientific research except for one thing: its motivation’. Very little of the Harwell work on fuel elements applied what was already known, yet all was aimed at an application: ‘the study of nuclear billiards inside metal crystals was applied research, because the motive was to develop a workable nuclear fuel element … but it was all applied science because its purpose was the development of nuclear power: … much deep, fundamental scientific research was done—and written up and published in learned scientific journals’. The interplay between new technology and new science is a theme of this paper: the applied sciences provide challenges that ask new questions of the basic sciences. Such interplay has long been evident in the semiconductor industries, to give one example. Another, earlier, example, would be the observation of thermodynamicist J. Willard Gibbs that thermodynamics owed more to the steam engine than the steam engine to thermodynamics.

Most of the examples I shall discuss relate to the UK Atomic Energy Authority (UKAEA), and especially to Harwell, the UK’s Atomic Energy Research Establishment. What follows is a very informal summary of the interplay between the science and technology of nuclear fission, and the way the scene evolved. Any historian might be embarrassed by my oversimplifications, but my aim is to say something about the research context during the key years of the development of fission in the UK. In emphasizing the role of Harwell, I am not trying to ignore the excellent and seminal work done in other laboratories.

It is very clear that national laboratories played an enormous role in the development of major new technologies. These laboratories, including both government and large industrial laboratories, were crucial to the achievement of nuclear energy. In many countries, they could provide the essential combinations of flexibility, continuity and integrated technology platforms that are hard to provide in other ways. Quite apart from big missions, like nuclear energy, such laboratories have had a remarkably positive effect on the prosperity of their region through the skills demanded by them or their suppliers. Indeed, such laboratories and programmes have proven to be infrastructure catalysts: Harwell and Culham spawned many high-tech firms in Oxfordshire, and the Inter-university Microelectronics Centre in Belgium has achieved much more than its original aim of improving the standards of the workforce in Flanders. Sadly, some governments have regarded such laboratories as costs, rather than a resource, and have wasted their potential (Stoneham 1996).

2. The nuclear experience: the evolution of the nuclear mission

The very first major nuclear mission was the atomic bomb. Much has been written about the technical successes of the Manhattan project, and even about the failures of the German programme. The urgency that World War II gave to nuclear weapon technology continued post-war, driven by the many changes and perceived threats in the world political scene. Some of the technologies developed for the military—such as isotope separation or radiation
detectors—were also fundamental to potential civilian uses, like electricity generation. Sometimes, civil applications, like electricity generation, had support from the military, since plutonium was a potential by-product. Security issues favoured separation of military matters from those that seemed solely peaceful, and a gradual separation of the two areas was natural. Thus, weapons work was devolved from Harwell to the Atomic Weapons Research Establishment at Aldermaston. By the late 1950s, it was possible for a scientist at Harwell to have a whole career without knowingly meeting another scientist working on weapons technology.

Electricity generation was the second big mission based on nuclear fission. This programme raised many technology questions, such as the choice of reactor design and of the management of large, novel projects in an emerging technology. The success of the UK programmes can be judged by the fact that the first reactor in western Europe (the Harwell GLEEP reactor) was switched on at the end of 1947, and the first nuclear power station producing commercial quantities of electricity (Calder Hall) was switched on in 1956; the last working reactor at Calder Hall was shut down only in 2003. Today’s time scales are unimpressive in comparison.

Still wider scientific issues emerged, such as basic ideas of radiation damage in component materials. Even in the 1950s, nuclear safety and radioactive waste were recognized issues. The 1957 Windscale reactor fire (Arnold 2007) provided a critical focus. Through some foresight in design, through the bravery and skill of the staff and through luck, there were essentially no casualties. Here, I might bring in a personal note since, at the time of the incident, I was at school in Barrow-in-Furness, some 20 miles from Windscale and the nearest large conurbation. My father, consultant obstetrician and gynaecologist for the population that would be most affected, and a very meticulous person, remarked to me several times that children born around that time showed no discernable evidence of radiation effects. Luck—in the weather—and simple measures, like throwing away milk, seem to have worked. Perhaps remarkably, those in Barrow at the time seemed remarkably relaxed about the incident. Whether this was because they had confidence in the Windscale staff, or because wartime experiences encouraged stoicism, I do not know.

While weapons and power were the primary missions, one should not forget the very important developments exploiting radioactive isotopes for medical and industrial uses. These have been enormously beneficial in saving lives. It must be added that the very large numbers of radioactive sources worldwide, and the low levels of supervision in some cases, have meant that the majority of serious radiation incidents have involved such sources, especially during disposal.

Clearly, the two major missions demanded skills that were not already available. These gaps were at all levels, from building and managing reactors to training apprentices. Major engineering projects had to be managed, and the understanding needed to get things right had to be developed. There was thus a research imperative. As a result, the 1954 Atomic Energy Act empowered the UKAEA to carry out research. This was wide-ranging because atomic energy was a new technology. A major consequence was the way the UKAEA supported British science, especially in its active involvement of universities (Keith 1993). In the period up to privatization, Harwell funded over 2000 PhD students and very many research projects. Moreover, UKAEA laboratories provided a
major infrastructure catalyst. Thus, very many of Oxfordshire’s hundreds of technology-based firms have involved staff (including technicians and apprentices) who were once at Harwell and Culham.

The strength of this science and technology base inevitably was recognized outside the areas clearly defined as atomic energy. However, UK industry was ambiguous in its attitudes. They were (sometimes) very happy to gain know-how and competitive new technologies—at least, if it was free—but they were horrified by the thought that work with government funding should ever compete with them. This led to legislation in 1965 that required the UKAEA to concentrate on UK industry, even when they did not want to know about it. Ion implantation, which transformed the semiconductor industry worldwide (with the possible exception of the UK, see Dickson 1985), illustrates this well. The technology was an imaginative development from particle-accelerator methods, but the UK semiconductor industry was un receptive, missing an opportunity and merely accelerating its own decline. Walter Marshall made a serious attempt to reposition the UKAEA to win more funding from industry through diversification. But by the early 1980s, government pressure and management practice were hinting at moves to privatization. Increasingly, large parts of the UKAEA were run as businesses, with all the associated problems but not always with the benefits. One particular gamble was the sacrifice of the research obligation to win essentially unrestricted freedom to address overseas markets. Atomic Energy Authority (AEA) technology came into independent existence in 1995.

3. Technical challenges and scientific consequences

Whenever there is urgency, as in a world war, it can drive collaborations that might be hard in peacetime. One example was the interplay between nuclear science and technology. But the demands of big programmes forced many other developments. Digital computing made possible the previously unachievable. It is no surprise that the earliest all solid-state computers were created at much the same time in the electronics industries and the nuclear laboratories. Nor is it surprising that these same organizations developed the first general-purpose material codes. At first, a new program was written for each new problem, perhaps using certain basic standard algorithms. But people like Frank Herman and Alan Lidiard realized that having a single, widely checked, code gave both speed and increased reliability.

Yet, not all the science needed heavyweight computing. Broadly, there are three levels at which theory can be important. At level 1, theory provides a framework to understand what is going on, a means to give system to observations and to seek likely ways forward. Examples would include the classic work on radiation damage by Kinchin & Pease (1955) and by Lindhard et al. (1963). At level 2, theory lets one scope a situation: sensibly accurate predictions tell one which factors are important and when they will be important. The analyses by Norgett et al. (1975) are still useful. Level 3 is reached when theory beats experiment. That might be because of time scales: one does not want to wait 100,000 years to test a solution to a radioactive-waste problem; one may not be able to observe events at the femtosecond time scale, irrespective

Phil. Trans. R. Soc. A (2010)
of their importance; there may be situations where the complexity or simply the scale of the phenomena mean that relative significance has to be identified by theory.

In the examples I shall give, there was a technological need for which scientific understanding led to practical solutions. These were just a few of the challenges, and do not describe the formidable engineering difficulties in creating a new industry on time and on budget. In addition to the new industry, there were scientific spin-offs: the science needed by the technology found its way into diverse other fields. In a field as broad as this, it is hard to do justice to everyone in references, and I shall usually refer to papers that themselves give extensive links to original sources.

(a) The challenge of voids

The primary defects expected in radiation damage are vacancies and interstitials. The natural expectation—at least at temperatures where at least one of these is mobile—is that most would simply recombine, restoring the perfect lattice. Others would be trapped at defects or impurities, those reaching the surface effectively disappearing and those reaching dislocations causing processes like climb. When there are high concentrations of vacancies and interstitials, they could aggregate, with dislocation loops having the lowest energy in a wide range of circumstances. It was therefore deeply disturbing scientifically to find voids, three-dimensional accumulations of vacancies, instead of the two-dimensional loops. It was still more disturbing technologically, since the voids were associated with significant swelling, both in structural materials and in fuel elements, and dimensional tolerances are tight in working reactors. The phenomenon is strongly temperature dependent and starts suddenly after an incubation dose, resulting in distortion of the core structures, unless controlled. Ultimately, the underlying science became clearer: there was a slight bias of some sinks for vacancies or interstitials, and the presence of He from α-particle production (i.e. transmutation was important) was able to alter the balance between two- and three-dimensional evolution at a critical stage. Perhaps more important in practice was the development of rate theory to describe the evolution of the mesostructure of these materials in a way that could be used alongside the practicalities of reactor engineering and operation (Brailsford & Bullough 1973, 1981). The science and technology developed together.

Voids showed yet another surprise. Evans (1971) showed that heavily-irradiated Mo contained a void lattice. The first such lattices comprised perhaps 10 million voids, each about 2 nm across, with a spacing of around 20 nm. The void lattice aligned with the host (body-centred cubic Mo) lattice, and even showed edge dislocations. The void lattice is just one of very many ordered structures, including shear planes, domains, flux lines and so on. Just how structures organize is discussed in a broader context by Stoneham (2007), who gives references to a wide range from the biological and physical sciences.

In the case of the void lattice, the main ideas group into two classes. One class suggests that void lattices are stable because of interactions between the voids, one important component being elastic interactions. The second class argues that void lattices are not stable, but form as the fastest growing instability, rather like spinodal decomposition. These ideas are not entirely exclusive, of course. There

Phil. Trans. R. Soc. A (2010)
are also many further suggestions, some based on very special mechanisms, e.g., assuming interstitials only move along specific planes or crystal axes. For the large-spacing lattices, it is very likely that the long-range elastic terms are crucial, and bias the voids during growth. Given the nature of the elastic interactions, I suspect that it is significant that the voids are never ideally spherical cavities. For the smaller bubble arrays, when there is plenty of He, for instance, growth instabilities might matter more, and indeed this is the fashionable explanation. However, growth instabilities do not explain the accuracy of the larger spaced lattices, or how the observed edge dislocations might form in the void lattice, or indeed why the lattice is so extended, not an ensemble of much smaller lattice-like regions.

The void lattice itself is of scientific interest, rather than technological significance. The formation of voids, however, was both a surprise and an unexpected phenomenon of considerable practical importance. It must be said, though voids were not entirely a surprise as the possibility had been raised at Harwell a few years earlier (Greenwood et al. 1959). This example is a reminder that we should not be scientifically complacent.

(b) The challenge of high temperatures

In many of the most serious hypothetical accident situations, oxide fuel is assumed to get very hot, possibly with uranium oxide fuel elements melting at above 3000 K. Thermal properties will be crucial in determining how the behaviour evolves, notably thermal conductivity and specific heat, but not forgetting mechanical properties. There is a need to consider issues like the fuel–coolant interaction when, in an extreme case, liquid UO\textsubscript{2} might contact a large volume of cooler water. Large-scale experiments plus insights from theory pointed to a complex sequence of events. First, the fuel would fragment. Steam would surround these fragments, reducing heat transfer to the cooler water dramatically. Serious problems would emerge should the steam layer be disrupted by a shock wave, which allowed a massive pressure buildup if water evaporation proceeded apace.

For a hypothetical accident scenario like this, there are at least three crucial roles for the underlying science. First, there are experiments on the system as a whole, like those done at the Molten Fuel Test Facility at Winfrith, UK. These experiments, individually, are costly, so a theory is needed that makes credible predictions possible for a far wider range of accident scenarios. But input has to include studies of UO\textsubscript{2} itself, and the ways that the thermal and even mechanical properties change dramatically above 2000°C. Understanding of the thermal properties needs to go significantly beyond the usual textbook ideas, partly from electronic terms. The most obvious electronic terms in the specific heat come from the uranium crystal field splittings. More important, and more interesting from a scientific point of view, is the fact that UO\textsubscript{2} is an intrinsic small-polaron semiconductor. ‘Small polaron’ means that the ionized electrons and holes are not the delocalized species found in metals or silicon, but are self-trapped because of the strong electron–phonon coupling (e.g. Stoneham et al. 2007). In the simplest ionic description, the hole h\textsuperscript{+} is a U\textsuperscript{5+} ion, and the electron e\textsuperscript{−} is a U\textsuperscript{3+} ion. The electron and hole move by incoherent hopping. Experimentally, it is found that they have mobilities close to the theoretical maximum, with one jump every...
lattice vibration period at high temperatures. The prediction of the numbers of electrons and holes at a given temperature lead to some of the first successful quantitative entropy calculations, using shell-model methods.

Polaron models imply definite dependences of properties on stoichiometry and on dopants. It is the case that the observed stoichiometry dependences do not always agree with what is observed, meaning some new ideas are still needed. Likewise, the origin is not understood of the apparent enhanced dislocation creep that causes mechanical properties to change dramatically at higher temperatures. Is this another case of recombination-enhanced diffusion, widely observed in semiconductor systems (Itoh & Stoneham 2001)? If so, what is the mechanism? Nonetheless, we have again a situation where basic sciences have been stretched by technological necessity, and have provided both a framework for the empirical studies and a solid base of understanding.

(c) The challenges of complexity of composition

If you have a system containing nearly 100 different chemical elements, and its composition changes with time, just how can you do technology? This is precisely the situation in many areas of nuclear technology, notably for spent fuel and its clad. Moreover, it is not just the composition that changes with time: the microstructure (grain structure) evolves substantially (e.g. Stoneham & Harding 2003). If one wanted to know the varieties of fission product species and how they are incorporated within the fuel or its microstructure, one could imagine doing a very extensive series of calculations at the state of the art. That would be a possible, albeit inefficient, way to identify which cases may cause problems. But that brute force approach (even if accurate) is not a serious way forward in some other cases. That route would be unhelpful if one wished to know how a UO₂-based nuclear fuel interacted with a zircaloy clad (both having complex microstructures), and how that interaction changes with time as the fuel is used up. Nor is a so-called first-principles approach likely to help if one wants to know how even simple properties—the density, say—of a glass storing high-level waste will change over a period of months or years.

When complexity of composition is an issue, this is the time for simple science-based models, possibly only to supply a framework for understanding experiments and their implications. As a first example, consider the interaction between an oxide and a metal. What causes adhesion? Here, one must be careful to ask which interface is considered. If the metal reacts with the oxide, a new oxide will form, and there will be two interfaces, one between the old oxide and the new, and one between the new oxide and the metal. Assuming that is sorted out, there are various systematic rules that are found in large databases of metal/oxide systems (Naidich 1981; Eustathopoulos et al. 1999). For quite a large number of interesting cases, the adhesion can be understood in terms of the image-charge model (Stoneham & Tasker 1985), in which the major part of the binding comes from the interaction of the charged ions in the oxide with the polarization (image) charges they create in the metal. This relatively simple picture has support from the experiment and from density functional theory, and successfully describes some tricky cases like anodic bonding and dependences on stoichiometry (e.g. Itoh & Stoneham 2001, §10.5). But even this model is too complex to do much more than give a framework of understanding for the interaction of a spent fuel with its clad.
Glasses are the usual solid-state media for immobilizing radioactive waste, since they will form a solid solution with most elements of the periodic table, they can be durable and they can be relatively easy to process. Issues include stability, solubility, tendency to form volatiles and possibly leach rates. What is needed are predictions over time scales long enough to satisfy social and political criteria. Those might reasonably be over times until the activity becomes less than that of the original ore, though the political need is closer to requiring an unlimited lifetime justified today. Those are some general issues, returned to later, but let us focus on radiation damage. Any realistic molecular dynamics would recognize the possible changes of charge states of ions, and the fact that even vacancies (though not simple to define in an amorphous system) can also change charge state, e.g. oxygen vacancies trapping electrons. Excitonic mechanisms can lead to ion displacements or enhance diffusion. There may well be long-range electric fields because electrons may leave the surface. Just because it is hard to define vacancies or interstitials in an amorphous system, where there is no reference perfect lattice, identifying displacements can be problematic. Structures can be analysed in different ways, and indeed approaches like reverse Monte Carlo can be informative. But what one wants usually is an enhanced analysis of behaviour and properties. For instance, what stored energy is possible, and what can be released if structural reorganization is possible when the glass gets hot? An example that illustrates the way a simple engineering question can challenge common approaches is how the density of a radwaste glass changes over months or years, and how this varies with composition. Some glasses expand with time, others contract. With so many chemical species, this is not a tempting problem for first-principles methods. What can be done, however, is (Stoneham 1993, following G. Tinnevella 1980, unpublished work) look at the wealth of data on glass densities as a function of composition. The rules describing the trends can (with some imagination) be used to predict a density for a radwaste glass. But the density of the glass coming from the vitrification plant has a slightly different density. A simple assumption might be that all the radiation does is speed up an approach to the rule-based density, i.e. what one might call radiation stirring. Interestingly, those simple ideas seem to fit all the glasses for which data are available.

(d) The challenge of radiation damage: beyond nuclear billiards

Cottrell referred to ‘the study of nuclear billiards’ in the 1950s, the early days of nuclear power. A decade later, Lindhard et al. (1963) identified systematic approximations that—up to a point—contain the essentials of the role of electrons in atomic collisions, i.e. the extent to which one must go beyond simple ‘billiard-ball’ models of the collision process. These important assumptions, which form the basis of most subsequent work, are as follows. First, electrons produced in collision cascades do not produce recoil atoms with appreciable energies. This is largely because electrons and nuclei have very different masses. In insulators and semiconductors, this assumption may fail. Secondly, atomic binding energies can be ignored for those heavy-particle collisions for which electronic stopping has appreciable significance. Thirdly, energy transfers from nuclei to individual electrons are relatively small. Fourthly, electronic and nuclear collisions contribute additively to energy loss, a superposition assumption that follows when there are only the many ‘weak’ collisions with electrons, often
with large impact parameters, if these scattered electrons are ineffective in causing ionic displacements. One can envisage cases where the effects are not additive, e.g. if the electrons cause a phase change, or if they cause cross-linking in a polymer.

Electron energy transfer to nuclei is well known. Energy transfer from nuclei to electrons is far less studied. Such transfers of energy between electrons and ions were discussed by Landau (1936) in the study of plasmas, but this was largely ignored until the work of Flynn & Averbach (1988). The consequences of transfer of energy from ions to electrons were surveyed by Stoneham (1989), identifying several distinct effects beyond the role of electrons in defining an interatomic potential. The several roles of electrons then became clearer (Stoneham 1989). Thus, the highly non-equilibrium electrons could act as a transient energy sink, taking energy that might lead to nuclear displacements. The excited electrons will redistribute energy in space on a scale similar to their mean free paths. The electrons will also damp the ionic motions in ways that are roughly viscous. Suppose an ion of mass $M$ enters a solid with velocity $V$ and energy $1/2(MV^2)$. The rate of energy loss is $AV$, where $A$ is a materials parameter depending on the electron density. If the initial kinetic energy is shared equally over $N$ ions, each ion has $1/N$ of the initial energy (velocity $v = V/N^{1/2}$), so the total rate of energy loss is $AVN^{1/2}$, more than that for the single ion. This has consequences seen in molecular dynamics. These several phenomena are discussed for metallic systems by Duffy (2010) and Itoh & Stoneham (2001) for non-metals.

Again, one fears complexity will make it hard to achieve useful results. But one should not despair. Many important results were obtained by Lindhard’s group, even before molecular dynamics and the like, based on analytical methods with simple ‘potentials’. Some significant results, at least, prove insensitive to the details of the potentials. Nonetheless, for quantitative predictions, it is important to understand just what potentials are and what they can do. Potentials are convenient functions that define a single energy surface as a function of ionic positions. Usually, potentials are chosen to be additive and transferable (it is not essential, merely convenient, to choose two-body, radial forms). If many energy surfaces are involved, as for many insulators, potentials will not work without generalization, even those based on so-called a priori methods that generate a single adiabatic surface. For instance, what would occur when a heavy ion deposits a large amount of energy—say 10eV per atom—in silicon? Silicon normally has an open structure, each atom covalently bonded to four others. The highly excited silicon presumably resembles liquid silicon, but with non-equilibrium electrons (and indeed some electrons may have left the highly excited region, so that it has a transient positive charge). But liquid silicon is metallic, with a higher density than the solid. So how might one model the evolution and recovery of this system?

A hierarchy of processes is a natural consequence of radiation. These hierarchies can be spatial, temporal or both. Thus, regarding time scales, there are fast processes, perhaps a few picoseconds, as conduction electrons eliminate electric fields and thermalize or as carriers in non-metals deform the lattice during self-trapping. There are steps on intermediate time scales of a few nanoseconds, such as luminescence, defect production or hot chemistry. Then there are slower processes, which are perhaps over the tens of years of a reactor lifetime, or
even geological times; such processes are often diffusion limited. Inevitably, the radiation response will often depend on history. Examples of history dependence would include changes following cross-linking in polymers or amorphization of a crystalline form. There may be sensitivity of the space distribution of primary damage to the nature of initial radiation and to other treatments, especially in situations where electric fields can build up to bias behaviour. Such electric fields can arise because of asymmetric irradiation or from trapping at mesoscale features. Examples include loss of electrons from a free surface, Compton scatter associated with the gamma flux in a Pb glass from the hot side of a post-irradiation damage cell, and the role of grain-boundary charge trapping.

Regarding length scales, there is much work on atomic-scale processes. These are the natural subject for molecular dynamics and full-scale electronic structure calculations. But one must remember that a grain of about 10 µm across will contain perhaps $10^{13}$ atoms, and the largest calculations handle perhaps 1 per cent of that number of atoms. At the engineering scale, finite-element methods are standard tools, widely used to exploit the classical continuum theories (elasticity, electromagnetism, thermodynamics), but needing material properties as input, whether from experiment or theory. The consequences of microstructure are crucial to bringing these components together. There is, in fact, a large history of calculations involving the mesoscale. Examples relevant to radiation damage include various forms of ablation and sputtering. It seems very rare for ‘multi-scale modelling’ (a large code that does calculations at more than one scale in the same run) to work well—the disadvantages of each method combine more easily than their merits (Stoneham & Harding 2003). It is very important to decide what is the question asked: the right method for one property may be wrong for others.

4. Technology challenges

(a) Atomic energy and its technologies

Nuclear technology for electricity generation has several aims. The system must produce electricity reliably, at acceptable cost, free from expensive regulatory restrictions both during its working life and afterwards, and generally acceptable to the population at large. These factors can be grouped, bearing in mind that the reactors are not all the same, but range from proposed new designs to existing reactors that are now quite old. For the reactors that are older, plant-life management is critical. When the Calder Hall reactors were built, economists did not expect any of them to last into the twenty-first century. The economists were wrong. Long lifetime for a facility that has a high initial capital cost has a substantial effect on electricity cost (Royal Academy of Engineering 2004).

Plant-life management involves monitoring the performance of operating reactors. One important component is post-irradiation examination of fuel elements removed from the reactor after some chosen level of burn-up. These elements are highly radioactive, and they must be examined by operators who are well shielded from their activity, and especially from the intense gamma emissions. Such examination needs clear viewing through windows that shield very well. Given the activity levels, the windows should be robust, lasting a
long time, since they are expensive and their replacement a difficult exercise. The gamma flux is not symmetrical, but directed outwards, and an interesting consequence is a ‘Compton wind’ that leads to electrons being scattered from the hot side towards the viewing side, a source of several problems that can be managed when understood.

Secondly, safety (and perhaps even more, safety as perceived by the public) has to be a major concern. What are the hypothetical accidents? How many levels of safety measures should be acceptable? Defence in depth proved effective for the safety of the public at Three Mile Island, despite a series of major errors. Are there new designs that can minimize—or even eliminate—major accidents? And how well can one check that the existing, working, structure does not have defects that could prove serious? It should be no surprise that the National Non-Destructive Testing Centre emerged within the AEA and with a focus on the new engineering designs and structures that demanded the highest levels of integrity.

Thirdly, is the spent fuel a mere waste (radwaste) or should it be reprocessed in some cycle to win the maximum energy from the original material? What technologies should be used if reprocessing is chosen? Reprocessing might have separate advantages if, for instance, it could enable the beneficial destruction of nuclear-weapon actinides. And if the decision is to avoid reprocessing, should one store the waste against some future actions, or attempt disposal? All of these questions presume that one can measure radiation effectively, both current activities and historic (accumulated) doses, and this leads to some interesting challenges.

(b) The challenge of plant-life management

How can a nuclear plant be operated safely, efficiently and economically, while minimizing interruptions for maintenance, and providing for longer plant life? The answer has to lie in combining a wealth of operating experience with a deep scientific understanding. Much of the relevant knowledge used directly in nuclear-plant design and operation concerns the engineering properties of materials, notably in-service changes in mechanical properties like fracture behaviour. Typically, these engineering properties might be hardness, toughness, brittleness and ductility. The modifications in behaviour from fast neutron exposures can dominate the behaviours central to safe plant operation of the plant. These altered macroscopic properties result from changes in the microstructure, which, in turn, stems from processes at the atomic scale. Knowledge of engineering properties demands detailed and evolving scientific understanding of damage processes, both basics and microchemical changes during in-service exposure. The range of time scales is huge (Stoneham et al. 1996), from $10^{-15}$ to $10^{-12}$ s for atomic processes to changes over plant operation lifetimes of years ($10^9$ s). Gaps remain, partly in fundamental topics, like the roles of electrons, but also in just how certain microscopic changes affect critical macroscopic properties such as fracture toughness. Plant-life management needs these links to be well established (Eyre & Matthews 1993). The conceptual framework exists, meaning that the engineering developments can go beyond simple empiricism. A deeper scientific understanding allows engineers to identify over-conservatism in the prediction of end-of-life properties.
The integrity of the reactor pressure vessel is crucial to the safe, economic operation of commercial light water reactors. The vessel is close to the reactor core, and gradually degrades mechanically owing to the relatively high flux of neutrons and γ-rays. The irradiation causes hardening and embrittlement, and specifically raises the ductile–brittle transition temperature, lowering the fracture toughness. These changes raise the probability of brittle failure during pressurized thermal shock transients, and so affect allowed pressure–temperature limits for start-up, shutdown and normal operation. Changes in these limits could affect the economic operation of the plant. A worldwide consensus (Phythian & English 1993; Odette 1995) has identified three main mechanisms controlling such embrittlement. First, radiation may create point-defect clusters and dislocation loops that cause matrix hardening. Secondly, grain-boundary segregation of embrittling elements like P may be enhanced by irradiation. Thirdly, there could be irradiation-enhanced formation of copper-rich precipitates in steels containing residual copper, leading to embrittlement at lower temperatures and doses. Further important issues include corrosion and stress corrosion cracking. Practical solutions may require very tight control of minor chemical constituents.

In each of these cases, phenomena on the atomic scale and extremely short time scales impact on the safe operation of plant over all its operational life. At the atomic scale, collisions of fast neutrons and lattice atoms can transfer energy from a few electron volts (eV) to tens of keV. The atom will be displaced if the energy transferred exceeds a threshold, \( E_d \), (typically above 40 eV), creating a vacancy and an interstitial. If the energy of the primary knock-on atom exceeds a few kiloelectron volts, a sequence of collisions causes a displacement cascade in a region typically less than 10 nm across. Here, at the centre of the cascade, considerable defect migration, clustering and annihilation may happen. Such cascades last for a few picoseconds, and are an important feature in fast neutron damage, as many recoils have energies above 30 keV. Since the natures and stabilities of the point defects created in these cascades control microstructural evolution and allied changes in material properties, a crucial radiation exposure parameter is the displacement per atom. Also crucial is the extent to which point defects in the cascade cluster. Small point-defect clusters within the cascade influence void swelling and irradiation hardening.

The cascades are so small and so short-lived that experiments concentrate on their debris, such as small surface craters, dislocation loops and stacking fault tetrahedra (Jenkins et al. 1993). However, a respectfully realistic picture of cascade evolution can be obtained though molecular dynamics, a case where the complementarity of theory and experiment is evident. Such theoretical studies help to establish an international standard for calculating displacements, as well as providing input to models of microstructure evolution. Norgett et al.'s (1975) approach (also incorporated into an American Society for Testing and Materials (ASTM) Standard E900; ASTM 1988) estimates the number of displaced atoms, \( N_d \), by partitioning energy between nuclear (displacement producing) and electronic losses, and uses a statistical process to evaluate the number of displacements resulting from the energy transferred in collisions. The result can be written \( N_d = kE_{\text{dam}}/2E_d \), where \( k \) is the damage efficiency factor, assumed energy independent with a value of 0.8; \( E_{\text{dam}} \) is the energy available for damage after energy loss to the electrons is taken into account and \( E_d \)
Review. Nuclear fission

is the mean displacement energy. This model now seems to overestimate the point-defect production in cascades. Subsequent computer simulations (Phythian et al. 1995) showed the damage efficiency to be energy dependent, in line with earlier experimental studies (Averback et al. 1978; Rehn & Okamoto 1987). Molecular dynamics show that the damage efficiency $k$ decreases from a value of about 1 at low energy to approximately 0.3 at energies above 1 keV, which can be extremely important for environments irradiated by fast electrons and fast neutrons.

Both molecular dynamics and electron microscopy give strong evidence for point-defect clustering in individual cascades. At doses for which cascade overlap occurs, some of the loops disappear in Ni, Cu and Ni–1%Al, and others change positions or Burgers vector or even coalesce with neighbouring loops as existing loops are engulfed by new cascades. Such overlap effects may be particularly important in components irradiated at relatively low temperatures, where there is little recovery and a dense microstructure develops. The evolution of the microstructures during fission neutron irradiation depends on those defects that survive within individual cascades, and also on the defect migration, which may involve a complex interaction with solutes. When both vacancies and interstitials are mobile, they can be trapped by interstitial impurities like C or N, or by dislocations. Radiation-induced segregation of alloy constituents is yet another factor contributing to complex microstructure.

A physical model is needed to describe how damage structures evolve. A successful approach has been the rate theory, developed originally to describe void swelling (Harkness & Li 1970; Bullough et al. 1975). This approach, which needs interstitial and vacancy concentrations as input, incorporates descriptions of phenomena such as solute segregation, irradiation creep and embrittlement. Thus, Stoller’s (1993a,b) ferritic steel embrittlement model identifies the implications of molecular dynamics for microstructure evolution and changes of mechanical properties under irradiation. This kinetic model uses reaction-rate theory to describe the formation and growth of point-defect clusters in neutron-irradiated pressure-vessel steels. Clusters impede dislocations and harden the matrix, increasing the yield strength, but leading to embrittlement. Such analyses show that defect production, clustering and subsequent hardening and embrittlement of the structural steel components depend in a complex way on irradiation dose and dose rate. Despite encouraging progress, linking microstructural and macroscopic properties is still difficult. It is not straightforward to predict dimensional changes, hardening, fracture toughness or corrosion from microstructural observations. Nor do increases in computer power make such quantitative understanding seem likely, though much can be learnt from large calculations. Thus, Becquart et al. (1993a,b) have used embedded atom potentials and molecular dynamics, predicting a critical toughness factor for the initiation of crack propagation. Prediction of mechanical property changes is harder, with the prediction of hardening most developed, but there are well-documented predictions of dimensional changes associated with void swelling.

The importance of these ideas may be seen from the practical example of how copper affects reactor pressure-vessel embrittlement. Here, the clearer understanding of atomic-scale processes made it possible to avoid over-conservative regulation. Matrix damage develops continuously during irradiation in the dose, dose rate and temperature range of interest, leading to hardening.
proportional to the square root of the dose (Phythian & English 1993; Odette 1995). Copper precipitation, however, rises to a plateau with irradiation, unchanged by subsequent irradiation, so the copper hardening reaches a plateau level when the dose rates are lower. In the reactor pressure vessel, steels with high levels of copper (greater than 0.25–3 wt%), the copper level in the matrix can differ significantly from the bulk value, since copper may be incorporated in second-phase particles during fabrication or heat treatment. Now it is solely the soluble Cu in the matrix at the start of life that controls embrittlement during operation. This matrix component must be measured to understand how embrittlement will proceed. The matrix copper level depends on the temperature of the final post-weld heat treatment. The lower the heat treatment, the lower the level of measured matrix copper. If welds have received a range of treatments, the regulatory guide may be unnecessarily conservative for those welds subject to a low post-weld heat treatment. In irradiated materials, measurement of matrix copper can establish whether the Cu precipitates have reached a plateau and the volume fraction of Cu available for further precipitation.

There is a clear technology gain from helping to establish appropriate regulation, not excessive regulation, and this illustrates one important role of scientific understanding. A parallel might be with the understanding of fracture and of dislocation dynamics and its impact on mechanical engineering and on non-destructive inspection. In addition, the approaches to radiation damage have proved a valuable base for assessing the next generations of reactors, including fusion reactors.

(c) The challenge of radioactive waste

Much of the present radioactive waste (‘radwaste’) came from early weapons programmes. For civil programmes, radwaste comes partly from nuclear fuel and its reprocessing, and also from decommissioning facilities. Ashby (2009) has rightly emphasized that, when a facility or product reaches its end of use, is it ‘waste’ or is there some residual value? In principle, there could be hybrid reactors that get nuclear energy from such waste, and there are schemes that propose to use weapons plutonium or $^{238}\text{U}$ as input fuel. Depleted uranium has proved formidable in munitions, where its density and mechanical properties (not its radioactivity, despite common misunderstandings) make it uniquely effective. The very careful analysis of possible roles of depleted uranium in the Gulf War and Balkan conflicts (Spratt et al. 2001, 2002a, b) largely ruled out links to the illnesses experienced by veterans (Jones et al. 2002; Wesseley & Freedman 2006). As regards the dangers of radioactive materials, one should note that the major radiological disasters over the last 60 years have arisen from improper disposal of medical sources, not from nuclear-power programmes (Nenot 2009).

Proposals for radwaste involve a decision between long-term storage (i.e. with access, should that be desired) or indefinite disposal. Different strategies are possible for low-level, medium-level and high-level wastes. How a particular radioactive species is treated will certainly depend on its half-life and the nature of its radiation. I shall not discuss the many complexities of the technical and political hurdles, but merely identify some issues in the immobilization of high-level waste in a glass or some solid form.
One major concern is that immobilized waste will be exposed to a flow of water that carries active species to the biosphere. Such waste might be vitrified high-level waste, for instance, incorporation in a glass form, glass being chosen partly because glasses can immobilize so many elements. The biosphere here means places where the nuclides might get into the food chain, for example. Some of the important issues are clearly site dependent, and choosing a very dry and geologically stable site is prudent. But what is the rate-determining step that determines how fast radioactive material gets into the water? What determines mass loss of the immobilized waste into the moving water?

The rate of mass loss of a radwaste glass, however, is not the same as its dissolution rate. Dissolution rates are what is measured when a radwaste glass is exposed to continually refreshed water, often in a Soxhlet experiment. The glass is usually supposed to dissolve congruently, so the material released into solution will match the average composition. This is almost certainly true for actinides and transition metals, at least the so-called network formers. It will not be true for He (from alphas) and is probably not true for network modifiers, like the lighter alkalis.

An important part of radioactive-waste management is making sure that mass loss is much slower than dissolution. This is done partly by making sure the water flow is minimal (‘dry’ conditions is the common descriptor, just as one might talk of putting on dry clothes) by choosing sites where there is little water and very little water flow. Partly this is done by backfill, which can both restrict flow and ensure conditions like pH are optimal. In relatively stagnant water, as in these dry conditions, there is redeposition as well as the dissolving of material. And the balance between dissolution and redeposition is roughly what determines the solubility. Solubility matters even more than dissolution under sensible flow conditions (Hughes et al. 1983), especially if one recalls that the radwaste is likely to be warm because of decays, so the solubility will diminish as the water moves to cooler regions away from the waste. Dissolved material will precipitate out, both reducing the amount present and blocking flow channels. Nonetheless, it is prudent to have low dissolution rates when practical.

There are two main groups of people at risk from radwaste, namely the public and the process workers who run the plant that vitrifies the radwaste. In most reasonable scenarios (and all that seem politically acceptable), the radiation workers are the more vulnerable. This means choosing processes that avoid high process temperatures that cause volatile components to evaporate. So a basic radwaste glass is chosen to have a low softening temperature. Such glasses often leach fairly easily, i.e. dissolution (while not fast) is not especially slow. There is thus a compromise, leading to strategies that aim first to prevent release from the initial containment (including the stainless steel drums and backfill), second, to ensure that, when release ultimately happens, it occurs as far as possible from people (‘people’ here would include those on any route that ends with people, e.g. via fish in the sea) and third, to ensure that, once release happens, there is effective dispersal so that no one person receives more than a small and ‘acceptable’ dose. No public enquiry is likely to accept primary containment as sufficient, although it is necessary. The second and third criteria are site specific. So radwaste containers should be placed in a geologically appropriate place with minimal water flow. Extrapolations to long times are tricky, partly because of changes of composition through radiation damage and
radioactive decays, and perhaps by the effects of radiation enhancement of some processes. Just what are long times can be misunderstood. An isotope with a one million year lifetime is, frankly, not seriously radioactive. One useful reference time is how long it takes for the waste to reach the activity level of the original ore. Experiment is crucial, but has its limits: laboratory experiments usually use high flow rates, and ‘real’ experiments with buried glass samples will only have time scales of a few tens of years. Archaeological vitrified material (including results from the Oklo phenomenon, the natural reactor in the French Gabon) involves glasses that have very different compositions, but can be used as a test of concepts and models.

5. Nuclear fission, past and future

It is increasingly accepted that there is no way to meet global climate change targets without substantial nuclear energy. The year 2007 was the first year in which more than half the world’s population lived in cities, and solutions for small isolated communities cannot be global solutions. There is optimism for fusion for the second half of the century, but fission seems essential before then. Moreover, the safety record of nuclear power in western Europe is exceptionally good, in terms of quantified health/mortality costs per kWh. Is there still the resource of experience and know-how to equip industry to take advantage of the opportunities? There is a new technology mission, but much of what was known has been lost, and some—like safety experiments needing large facilities—potentially expensive to recreate. Certainly, computer-based methods are a much more formidable resource today. Certainly, there will be much novel basic and applied research to be done. While much of this research can be done in universities, it will be far harder to carry out large-scale technological programmes because of the loss of national laboratories since the 1980s (Stoneham 1996).

There has been some attempt to recover the situation in preparation for the building of a new generation of nuclear power stations in the UK. Since 2005, there has been a series of measures to support university research and to provide technical education for nuclear engineering. Part of this process has been the provision of some larger university-based facilities for technology development in waste immobilization, decommissioning, advanced manufacturing and reactor technology. The National Nuclear Laboratory was set up in 2008 to establish a national capability from the research arm of British Nuclear Fuels plc (BNFL), including parts of the former AEA and Central Electricity Generating Board (CEGB) capabilities and the Sellafield Technology Centre. The National Nuclear Laboratory will be dependent on contracts from the nuclear industry and the Government regulators for its income. The limited supply of uranium at acceptable cost (the immense amount in low-grade ores and in the sea is expensive to capture) makes it necessary to seek a more efficient fuel cycle and reactor systems that can generate new fuel from fertile materials like $^{238}$U and $^{232}$Th. There will be a Nuclear Centre of Excellence that will look at the challenge of making fuel cycles proliferation proof (Brown 2009), but the challenge to design and build a new fourth-generation reactor system needs to be addressed now if fission power is to continue beyond 2050.
While this present article is about peaceful uses of nuclear energy, atomic weapons have been a conspicuous component of the world scene for a long time. Just as the technical thrust for civil nuclear power has changed over the years, so the aims of weapons technology have evolved, at least in those countries that have had weapons for some time. Efforts are less aimed at developing new weapons than on avoiding testing and on the safety and reliability of existing weapons, i.e. making sure they would work only if required to do so. The move has been to substitute testing by modelling, and this itself requires a substantial and effective understanding of radiation effects.

The fusion reactor lies outside my present scope (Duffy 2010), but current programmes are identifying many materials challenges, analogous to those developing fission reactors met in the 1950s. There is a need to rethink key materials: there are special demands for the divertor, there are possible problems of dust and of degradation of insulators; the breeding of tritium must be done efficiently. How much of the structural material needs to be ductile? And to evaluate these materials realistically, novel radiation sources are needed, with proper neutron energies, dose rates and He production.

The main theme of this paper has been the interplay of science and technology. This interplay was central to the early successful developments over half a century ago. Such interplay has continued in a key way to keep nuclear power as a significant fraction of electric power in many parts of the world, and certainly an extremely safe source in western Europe. In the next half century, it is hard to see how demands for electricity can be met without expansions of nuclear fission power stations. The acceptability of these reactors will rely on a further period in which new technologies and new science work together.

I am indebted to Professor J. R. Matthews and Dr Dorothy Duffy for valuable discussions and comments.

References


Phil. Trans. R. Soc. A (2010)


Keith, S. T. 1993 *The fundamental nucleus: a study of the impact of the British atomic energy project on basic research*. Harwell, UK: AEA Technology


